

Constraining the equation of state of nuclear matter from competition of fusion and quasi-fission in the reactions leading to production of the superheavy elements

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Abstract

The mechanism of fusion hindrance, an effect preventing the synthesis of superheavy elements in the reactions of cold and hot fusion, is investigated using the Boltzmann-Uehling-Uhlenbeck equation, where Coulomb interaction is introduced. A strong sensitivity is observed both to the modulus of incompressibility of symmetric nuclear matter, controlling the competition of surface tension and Coulomb repulsion, and to the stiffness of the density-dependence of symmetry energy, influencing the formation of the neck prior to scission. The experimental fusion probabilities were for the first time used to derive constraints on the nuclear equation of state. A strict constraint on the modulus of incompressibility of nuclear matter $K_0 = 240 - 260$ MeV is obtained while the stiff density-dependences of the symmetry energy ($\gamma > 1.$) are rejected.

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In the last two decades of the past century, the heavy elements up to $Z=112$ were synthesized using cold fusion reactions with Pb, Bi targets in the evaporation channel with emission of one neutron [1]. The experimentalists had to face a rapid decrease of cross sections down to the picobarn level due to increasing fusion hindrance whose origin was unclear. Since the turn of millennium, still heavier elements with $Z=113-118$ were produced in the hot fusion reactions with emission of 3-4 neutrons using ^{48}Ca beams with heavy actinide targets between uranium and californium [2–8]. Again the increase of fusion hindrance was observed, caused by competition of the fusion process with an alternative process called quasi-fission. Quasi-fission occurs when instead of fusion the system forms elongated shape evolves towards the scission point. The systematics in the reactions with lead target, published in [9], shows that quasi-fission sets on for beams ^{48}Ca and heavier. In terms of reaction mechanism, quasi-fission is similar to nucleon exchange between colliding nuclei, however it proceeds while the shape of the system also changes dramatically. Compared to the fusion-fission, proceeding via formation of compound nucleus, angular distribution of fission fragments is forward-peaked in center-of-mass frame, total kinetic energy (TKE) is lower and the mass asymmetry ranges from the mass asymmetry of projectile-target configuration towards the symmetric mass split, with the yield decreasing monotonously. A large systematics of high quality data on quasi-fission in the reactions, leading to production of superheavy elements, was obtained in the recent years in Dubna [10] and Tokai [11]. It is usually considered that the process of quasi-fission is governed by a complex dynamics of the projectile-target system, which is often described using theoretical tools such as the model of di-nuclear system [12] or the Langevin equation [13, 14]. Besides the above theoretical tools, the competition of fusion and quasi-fission was also addressed using the implementations of the Boltzmann equation known as ImQMD [15] and using the time-dependent Hartree-Fock theory [16]. However, success of a simple statistical model of fusion hindrance, introduced in [17, 18] suggests that the competition of fusion and quasi-fission could be dominantly driven by the available phase-space and hindrances originating in diabatic dynamics are not decisive. In the present work we employ the Boltzmann-Uehling-Uhlenbeck (BUU) equation with the Pauli principle implemented separately for neutrons and protons and with the Coulomb interaction. We demonstrate how various equations of state of nuclear matter implemented into such transport simulation influence the competition of fusion and quasi-fission. Based on available data on reactions, leading to production of super-heavy nuclei, we extract most stringent constraints on the stiffness of the nuclear equation of state and on the density-dependence of the symmetry energy.

In order to describe theoretically the competition of fusion and quasi-fission at energies close to the Coulomb barrier, the goal is to describe the evolution of the nuclear mean field of the two reaction partners. However, besides nuclear mean field it is necessary to take into account the electrostatic interaction among protons and also it is necessary to guarantee preservation of the Pauli principle in a strict way. The evolution of nuclear mean field can be described by solving the Boltzmann equation. One of the approximations for the solution of the Boltzmann equation, the Boltzmann-Uehling-Uhlenbeck model is extensively used [20, 21], which takes both the nuclear mean field and the Fermionic Pauli blocking into consideration. The BUU equation reads

$$\frac{\partial f}{\partial t} + v \cdot \nabla_r f - \nabla_r U \cdot \nabla_p f = \frac{4}{(2\pi)^3} \int d^3p_2 d^3p_3 d\Omega$$

$$\frac{d\sigma_{NN}}{d\Omega} v_{12} \times [f_3 f_4 (1-f)(1-f_2) - f f_2 (1-f_3)(1-f_4)] \delta^3(p+p_2-p_3-p_4), \quad (1)$$

where $f=f(r, p, t)$ is the phase-space distribution function. It is solved with the test particle method of Wong [22], with the collision term as introduced by Cugnon et al. [23]. In Eq.(1), $\frac{d\sigma_{NN}}{d\Omega}$ and v_{12} are in-medium nucleon-nucleon cross section and relative velocity for the colliding nucleons, respectively, and U is the sum of the simple single-particle mean field potential with the isospin-dependent symmetry energy term

$$U = a\rho + b\rho^\kappa + 2a_s\left(\frac{\rho}{\rho_0}\right)^\gamma \tau_z I, \quad (2)$$

where $I = (\rho_n - \rho_p)/\rho$, ρ_0 is the normal nuclear matter density; ρ , ρ_n , and ρ_p are the nucleon, neutron and proton densities, respectively; τ_z assumes the value 1 for neutron and -1 for proton, the coefficients a , b and exponent κ represent the properties of symmetric nuclear matter, while the last term describes the influence of the symmetry energy, where a_s represents the symmetry energy at saturation density and the exponent γ describes the density dependence.

The in-medium nucleon-nucleon cross sections are typically approximated using the experimental cross sections of free nucleons (e.g. using the parametrization from Cugnon [23]). Alternatively, as shown in the work [24], in-medium nucleon-nucleon cross sections can be estimated directly using the equation of state and used successfully e.g. to describe the evolution of transverse flow in a wide range of relativistic energies [19, 25]. However, at low energies close to the Coulomb barrier the collision term plays only limited role and the choice of the in-medium nucleon-nucleon cross sections does not influence the results of simulations, in part because at such low relative momenta both cross sections exceed the cutoff value applied in the BUU code.

In order to describe the nuclear collisions close to the Coulomb barrier, it is crucial to implement properly the electrostatic interaction among protons. It is however impossible to introduce a density-dependent term into the single-particle potential in the equation (1), since electrostatic interaction has a long-range and for the infinite nuclear matter it would diverge. In the present work, instead of modification of the single-particle potential, we complement the corresponding density-dependent force $\nabla_r U$ acting at a given cell of the cubic grid (with a mesh of 1 fm) with the summary force generated by the proton distribution outside of the cell. This approach thus avoids fluctuation of the Coulomb force due to interaction of protons inside the cell, what is natural since this interaction is considered in the collision term. Specifically we consider only interaction with the protons of the same set of test particles, which allows to perform simulations practically with the same CPU time consumption as the simulations without Coulomb interaction. This circumstance allowed to perform this study in principle. Besides the introduction of Coulomb interaction, at low beam energies close to the Coulomb barrier it is necessary to guarantee strict preservation of the Pauli principle. We assure this by implementing the Pauli principle separately for protons and neutrons.

The simulations were performed using various assumptions on the stiffness of the equation of state of symmetric nuclear matter, as represented by the single-particle potential in Eq. (2). The exponent κ was varied between values of 7/6 and 2, corresponding to the range of incompressibilities between 200 and 380 MeV (the value of incompressibility depends linearly on κ). Besides the stiffness of the equation of state of symmetric nuclear matter, we

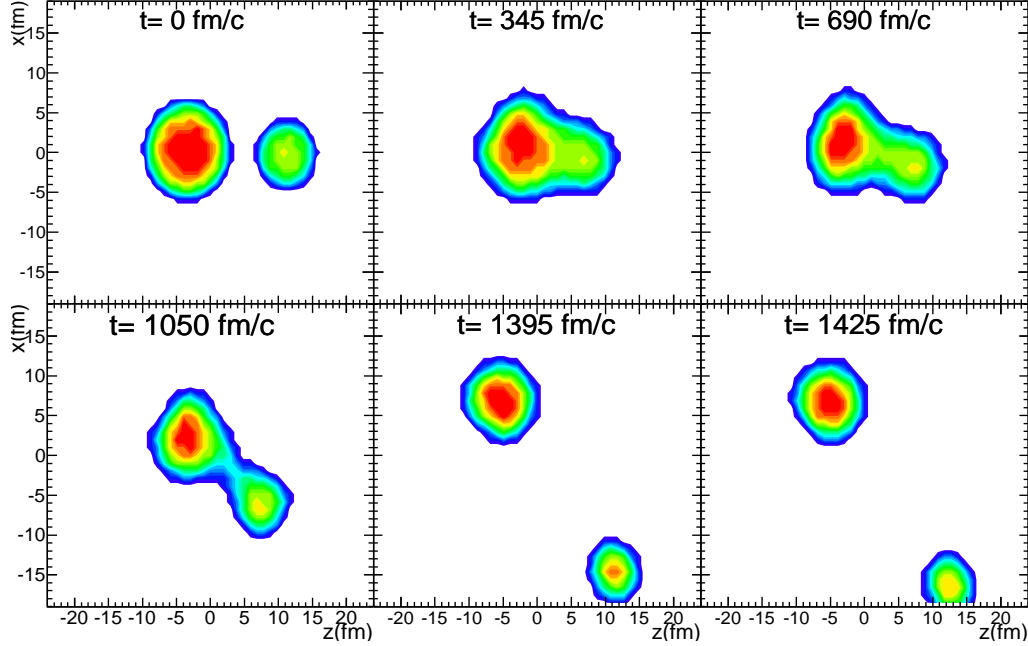


FIG. 1: Typical evolution of nucleonic density for the central collision $^{64}\text{Ni}+^{186}\text{W}$ at beam energy 5 AMeV, simulated using the soft equation of state with incompressibility $K_0 = 202$ MeV and the soft density dependence symmetry energy with $\gamma = 0.5$. Weak surface tension is overcome by Coulomb interaction and quasi-fission occurs.

implemented several assumptions on the stiffness of the density dependence of symmetry energy by varying the exponent γ in Eq. (2) between 0.5 and 1.5. For each calculated reaction, the simulation was performed using 600 test particles, with 20 different sequences of the pseudo-random numbers. The simulations were performed using a computing workstation with four Xeon Phi coprocessor cards with 61 cores, allowing to perform hundreds of simulations (up to one thousand) in parallel.

In order to investigate the role of the equation of state of nuclear matter in the competition of fusion and quasi-fission in reactions leading to heavy and superheavy nuclei, we selected a representative set of reactions, where experimental data exists. As one of the heaviest systems, where fusion is still dominant, we use the reaction $^{48}\text{Ca}+^{208}\text{Pb}$. This reaction was measured [26, 27], and a typical dominant peak at symmetric fission was observed in the mass vs TKE spectra of fission fragments, with TKE consistent to fusion-fission proceeding through formation of the compound nucleus ^{256}Nb . Onset of quasi-fission was observed [28] in the reaction $^{64}\text{Ni}+^{186}\text{W}$, leading to compound system ^{250}No , where a prominent fusion-like peak is not observed anymore, however symmetric fission, which can be attributed to fusion-fission, is still observed relatively frequently. Quasi-fission becomes even more dominant in the reaction $^{48}\text{Ca}+^{238}\text{U}$, nominally leading to compound nucleus ^{286}Cn . Nevertheless, the symmetric fission events still amount to about 10 % of fission events [29]. Comparison with the reaction $^{64}\text{Ni}+^{186}\text{W}$ shows that the relative amount of symmetric events in reaction $^{48}\text{Ca}+^{238}\text{U}$ is twice lower than in the reaction $^{64}\text{Ni}+^{186}\text{W}$, thus implying the relative amount of 20 % of symmetric fission for the latter reaction. In reactions $^{64}\text{Ni}+^{208}\text{Pb}$ [26], $^{48}\text{Ca}+^{249}\text{Cf}$ [7], and $^{64}\text{Ni}+^{238}\text{U}$ [30] the quasi-fission already dominates and fusion hindrance amounts to

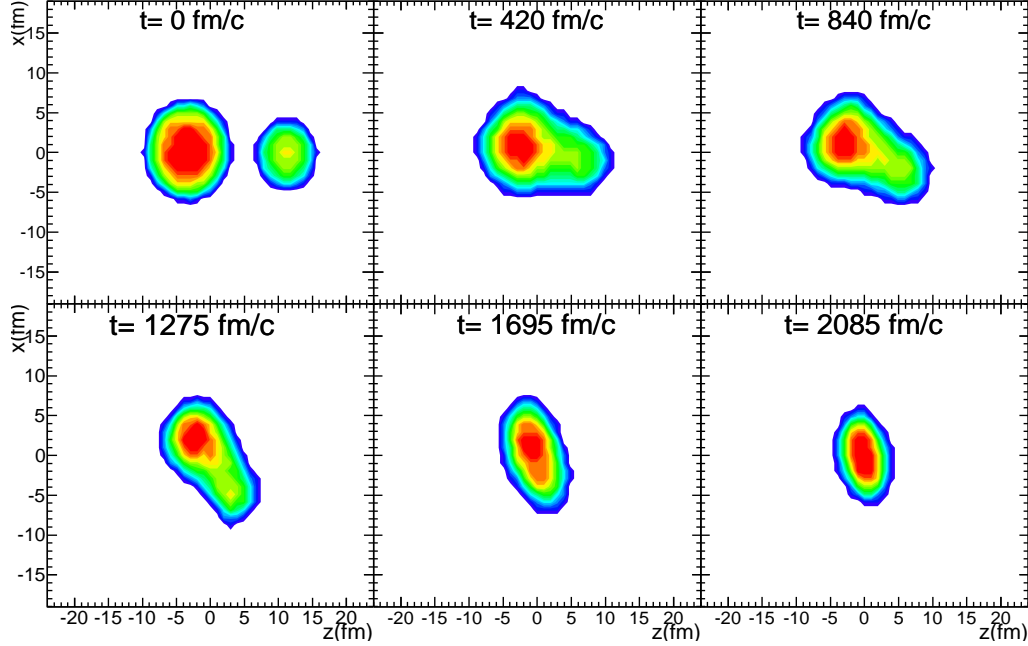


FIG. 2: Typical evolution of nucleonic density for the central collision $^{64}\text{Ni}+^{186}\text{W}$ at beam energy 5 AMeV, simulated using the stiff equation of state ($K_0 = 300$ MeV) and the soft density-dependence of symmetry energy ($\gamma = 0.5$). Stronger surface tension overcomes Coulomb interaction and quasi-fission is prevented.

several orders of magnitude ($10^{-3} - 10^{-5}$ [17, 18]).

Simulations were performed at beam energy 5 AMeV, which is above the Coulomb barrier and in all cases corresponds to the nearest experimental point within few MeV. Since the angular momentum range where quasi-fission events are produced is not known precisely and also to assure that we won't observe deep-inelastic transfer, which occurs at peripheral collisions, we simulate the most central events, with impact parameter set to 0.5 fm (exactly central events practically do not occur in experiment). Simulations were performed up to the time 3000 fm/c, sufficient for formation of the final configuration in all investigated cases. To carry out comparison with experiment, we need to determine from available experimental information the probability of fusion in central collisions. For the reaction $^{48}\text{Ca}+^{208}\text{Pb}$ the fusion probability is close to 100 %, while for reactions $^{64}\text{Ni}+^{208}\text{Pb}$, $^{48}\text{Ca}+^{249}\text{Cf}$, and $^{64}\text{Ni}+^{238}\text{U}$ it is close to zero ($10^{-3} - 10^{-5}$). Of the two remaining reactions, the total fusion probability of 10 % and the fact that fusion probability peaks at central collisions infer the constraint on fusion probability in the reaction $^{48}\text{Ca}+^{238}\text{U}$ at central events between 20 - 50 % (upper limit is based on assumption that quasi-fission is dominant even in central collisions). Since comparison of shapes of experimental mass distribution in reactions $^{48}\text{Ca}+^{238}\text{U}$ and $^{64}\text{Ni}+^{186}\text{W}$ shows that there is approximately twice higher relative abundance of fusion in reaction $^{64}\text{Ni}+^{186}\text{W}$, we constrain the fusion probability in this reaction at central collisions between 40 - 80 %. These constraints remain still relatively loose, but they reflect the dominant scenarios and thus the dynamics of competition of fusion and quasi-fission. As a consequence, this representative set of reactions allows to constrain the parameters of equation of state of nuclear matter.

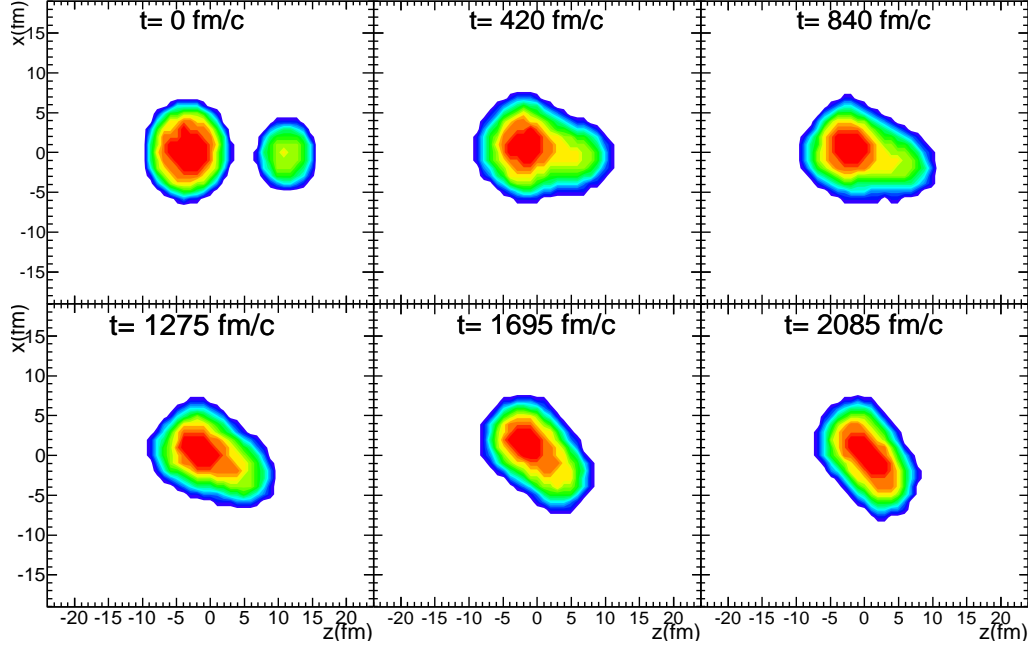


FIG. 3: Typical evolution of nucleonic density for the central collision $^{64}\text{Ni}+^{186}\text{W}$ at beam energy 5 AMeV, simulated using the soft equation of state ($K_0 = 202$ MeV) and the stiff density-dependence of symmetry energy ($\gamma = 1.5$). Despite weak surface tension the stiff density-dependence of symmetry energy prevents formation of a neck and quasi-fission is prevented.

From the investigated reactions, the collisions of $^{64}\text{Ni}+^{186}\text{W}$ exhibit highest sensitivity to the parameters of the equation of state. Fig. 1 shows typical evolution of nucleonic density for the collision $^{64}\text{Ni}+^{186}\text{W}$, simulated using the soft equation of state with incompressibility $K_0 = 202$ MeV and the soft density-dependence of symmetry energy $\gamma = 0.5$. One can see that the impinging projectile nucleus establishes contact with the target nucleus, however the weak surface tension, caused by the soft equation of state, is not sufficient to overcome Coulomb repulsion of the projectile and target which re-separate after approximately 1200 fm/c (scission time is comparable with other approaches [12–16]). Similar evolution was observed in all 20 simulated test particle sets. Fig. 2 shows simulation of the same reaction with $K_0 = 300$ MeV and $\gamma = 0.5$. In all simulations of this case the stronger surface tension generated by the stiff equation of state allows to overcome the Coulomb repulsion and system undergoes fusion. Strong sensitivity to the stiffness of the equation of state is thus demonstrated. Fig. 3 shows the simulation with $K_0 = 202$ MeV and $\gamma = 1.5$. One can observe that increased stiffness of the density-dependence of symmetry energy can also prevent system from separating into two fragments. In this case the weak surface tension allows to form elongated configuration (similar to Fig. 1), however the stiffer symmetry energy prevents formation of a low-density neutron-rich neck and thus the contact between the two reaction partners is preserved until the surface tension finally overcomes the Coulomb repulsion. Figs. 1 - 3 demonstrate a strong sensitivity of the system $^{64}\text{Ni}+^{186}\text{W}$ to the parameters of the equation of state.

In the other system of comparable mass, collisions of $^{48}\text{Ca}+^{208}\text{Pb}$ typically result in fusion, with exception of the simulations with $K_0 = 202 - 230$ MeV and $\gamma = 0.5 - 1.0$. For such soft

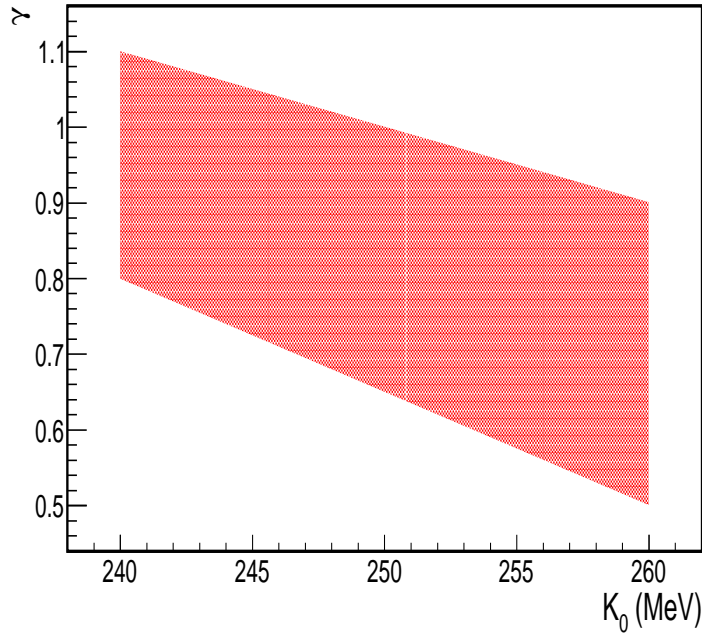


FIG. 4: Constraint on stiffness of symmetric nuclear matter (modulus of incompressibility) and on density-dependence of the symmetry energy (exponent γ from Eq. 2) derived from the simulations of competition between fusion and quasi-fission.

equation of state collisions usually result in quasi-fission and thus such equations of state can be considered in conflict with experiment. Heavier systems $^{64}\text{Ni}+^{208}\text{Pb}$, $^{48}\text{Ca}+^{249}\text{Cf}$, and $^{64}\text{Ni}+^{238}\text{U}$ usually undergo quasi-fission for $K_0 = 202 - 255$ MeV, for stiffer equations of state fusion appears and eventually becomes dominant. Thus a stiff equation of state with $K_0 = 272 - 300$ MeV can be rejected. Also a stiff symmetry energy with $\gamma = 1.5$ combined with soft equations of state with $K_0 = 202 - 255$ MeV lead to fusion and thus can be rejected. The remaining system $^{48}\text{Ca}+^{238}\text{U}$ behaves similarly to $^{64}\text{Ni}+^{186}\text{W}$, consistently with constraints derived from other systems.

As a result of the analysis of competition between fusion and quasi-fission, it was possible to set a rather strict constraint on the incompressibility of the equation of state of nuclear matter $K_0 = 240 - 260$ MeV with softer density dependence of the symmetry energy with $\gamma = 0.5 - 1.0$ (see Fig. 4). This constraint is based on simulations of collisions, where maximum density reaches 1.4 - 1.5 times the saturation density. The shape of the constrained area reflects a trend of softening the density-dependence of the symmetry energy, necessary to balance increase of incompressibility. Such trend stems from competition of the surface tension, related to the stiffness of the equation of state of symmetric nuclear matter, with the Coulomb repulsion. This corresponds to the traditional picture of nuclear fission, where fissility of the system is defined as a ratio of the Coulomb repulsion to twice the surface energy. However, the present analysis goes beyond this simple macroscopic picture and elucidates the crucial role of the density-dependence of the symmetry energy in the dynamics of the system close to the scission point. In comparison with other methods, such as constraining the equation of state using the nuclear giant resonances [31–33] or the flow

observables in relativistic nucleus-nucleus collisions [34], in the present analysis the effect of nuclear equation of state is manifested directly, and thus it is not affected by uncertainty related e.g. to description of underlying nuclear structure in the former or disentangling the effect of the two-body dissipation via nucleon-nucleon collisions in the latter case. In our simulations the microscopic nuclear shell structure is not considered. Role of the shell structure in nuclear fission remains an open question, as demonstrated by the recent observation of asymmetric fission of ^{180}Hg [35], contrary to expectations, based on the shell structure of fission fragments. The effect of closed nuclear shells can be manifested differently in fusion and quasi-fission channel, specifically for each system, and thus no simple trend must necessarily exist. By using a representative set of investigated reactions we provide a solid base for assumption that the extracted constraints do not depend critically on the effect of shell structure. In order to obtain even more strict constraints, in particular on the density-dependence of symmetry energy, more experimental data are necessary close to the onset of quasi-fission, where the sensitivity of the neck dynamics to the density dependence of the symmetry energy is highest.

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